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Ultracold Polar Molecules: New Phases of Matter for Quantum Information and Quantum Control

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14. ABSTRACT
In this first funding period we have obtained several key results towards the control of cold collisions between molecules, and the prediction and understanding of exotic many-body phases of ultracold molecules trapped in optical lattices. Highlights are: (a) The computation of the elastic and reactive rates of quasi-two-dimensional and quasi one-dimensional collisions of highly reactive ultracold molecules (in collaboration with the UMd MURI PI Paul Julienne). Some of these predictions have been now verified experimentally by the JILA group [deM11]. (b) The determination of the phase diagram of ultracold polar molecules trapped in optical lattices in two [Cap10] and one [Dal10] dimensions, showing that the groundstate of the system can support exotic phases such as supersolid phases and Devil-like's staircases of lattice solids [Cap10, Dal10]. (c) The study of the physics of dipolar gases as can be realized in novel setups, such as quantum gases of laser-dressed atoms [Pup10, Sch10]. These works have already resulted in several high-profile publications in this first funding period. Future work will focus on the analysis of the quantum phases of bosonic and fermionic polar molecules in multi-layer structured, as are being realized in experiments with molecules trapped in several layers of an optical lattice. We expect that several interesting pairing mechanisms will arise in these systems, possibly leading to many-body phases of interest. In addition, we will work towards the realization of new architectures for quantum computing with polar molecules which make specific use of long-range interactions in these systems.

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Ultracold polar molecules: new phases of matter for quantum information and quantum control, in response to ONR BAA Announcement Number 08-019, Research Opportunity (12): Ultracold Molecules. PI: Guido Pupillo, co-PI: Peter Zoller

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1. Summary

The goal of this proposal is to support the work of the UMd MURI project in understanding and exploiting the electric, magnetic and electromagnetic field control of ultracold molecules and to use the novel control features of these systems to produce and characterize new phases of quantum matter. In Innsbruck, we aim at bringing together: (i) the microscopic physics of cold collisions of the molecules, in the context of the realization of collisionally stable setups, (ii) the macroscopic physics of the study of the quantum phases which can be realized with strongly interacting polar molecules, and (iii) the exploration of quantum optics and quantum computing applications of lattices of polar molecules. A key goal is to work in strong collaboration with the theory and experimental groups of the UMd MURI team to achieve the experimental realization of systems where molecules can collide with suppressed losses.

In this first funding period we have obtained key results on the control of cold collisions between molecules, and the many-body phases of ultracold molecules trapped in optical lattices, reaching substantial progress on several of the points listed above. These works resulted in several high-profile publications in international journals (see *List of published papers within EOARD*). Here are a

few highlights: (a) In a collaboration with the UMd-MURI PI Paul Julienne [Mic10], we have made specific predictions for the elastic and reactive rates of quasi-two-dimensional and quasi one-dimensional collisions of highly reactive ultracold molecules, predicting that evaporative cooling should be achievable in experiments with KRb molecules performed at JILA. Some of these predictions have been now verified experimentally by the JILA group [deM11]. (b) We have determined the phase diagram of ultracold polar molecules trapped in optical lattices in two [Cap10] and one [Dal10] dimensions, showing that the groundstate of the system can support exotic phases such as supersolid phases, corresponding to the coexistence of solid and superfluid orders, and so-called Devil's staircases of lattice solids [Cap10, Dal10]. The supersolid phase that we have demonstrated in [Cap10] corresponds to the first microscopic description of such a state. (c) We have opened a new line of research in the study of dipolar gases by proposing ways to obtain long-range interactions in quantum gases of ground-state atoms [Pup10, Sch10] weakly-dressed by laser light. We have shown that under realistic experimental conditions several possible quantum phases and phase transitions can be realized in these systems, including a quantum phase transition to a supersolid in free-space [Cin10].

2. Introduction

The availability of ultracold atoms in gases and lattices, together with theories for understanding and predicting their properties, has become a transformational multidisciplinary area of contemporary science. The goal of the UMd MURI proposal is to extend this work to ultracold molecules by integrating state-of-the-art experiment and theory in order to expand ultracold matter research into new domains of forefront science of interest to the Department of Defense. The existence of strong dipolar forces between polar molecules offers new scientific prospects that are not available with ultracold atoms. The UMd MURI proposal comprehensively addresses all of the key experimental and theoretical aspects of making and using ultracold molecules in the microkelvin domain. The success of the proposal requires combining experimental and theoretical efforts in atomic, molecular, and optical physics, few-body physics, the condensed matter physics of quantum degenerate atomic and dipolar molecular gases, quantum chemistry, and quantum information science.

In our work in Innsbruck we bring together the microscopic physics of cold collisions of the molecules, in the context of the realization of collisionally stable setups, and the macroscopic physics of the study of the quantum phases which can be realized with strongly interacting polar molecules. The core of our work in support of the UMd MURI proposal has thus a markedly interdisciplinary character, which aims to help bring together the various aspects of the few-body and many-body physics of interacting polar molecules.

In this first funding period, we have worked in close connection with theorists and experimentalists in the UMd MURI team, and in particular with the UMd MURI PI Paul Julienne and the JILA group. This effort has resulted in a detailed study of collisional rates for reactive KRb molecules, where we have shown that inelastic rates can be efficiently suppressed by confining particles to a 2D geometry using an optical lattice combined with an external polarizing field [Mic10]. Experimentalists at JILA have recently implemented this scheme in a ground-breaking experiment published in Nature

Physics [deM11]. Following our prediction, they are now trying to achieve evaporative cooling of the molecules in 2D geometry, which would constitute a giant step in the field. In addition, we have proposed and analyzed the quantum phases that will be realized in experiments with ultracold fermionic and bosonic molecular gases. In particular, we have analyzed the possibility to realize exotic quantum many-body phases in several different physical setups, ranging from ultracold molecules trapped in one and two-dimensional lattices [Cap10,Dal10], to quantum gases of Rydberg-dressed ground-state alkali atoms [Pup10,Cin10,Sch10]. These results constitute significant progress towards the completion of the goals of this EOARD proposal.

We are currently working on the possibility to realize a novel quantum computing architecture which directly exploits the long-range nature and the tunability of the dipole-dipole interactions of cold polar molecules.

In the following we detail key results that we have obtained in this first funding period and we related them to the proposed research subjects. These include: the study of stability and control of polar molecules (Sect.3), new phases of quantum matter (Sect.4), and the proposal of novel setups and methods for studying dipolar physics in ultracold gases (Sect.5).

3. Ground state polar molecules: collisional stability

The JILA-Exp group has a source of about 40000 vibrational ground state KRb molecules near 350 nK and is on the verge of producing a quantum degenerate gas of stable polar molecules for a range of exciting applications, including studies of new forms of quantum matter, quantum phase transitions, and other collective quantum effects. The UMd MURI and EOARD proposal has as a key goal to understand the novel ultracold chemistry and the field control possibilities of these molecules, and to put them into reduced dimensional optical lattice structures in order to realize new phenomena with quantum matter or to realize new architectures for quantum information applications. In the following we detail our progress and results towards the completion of these goals.

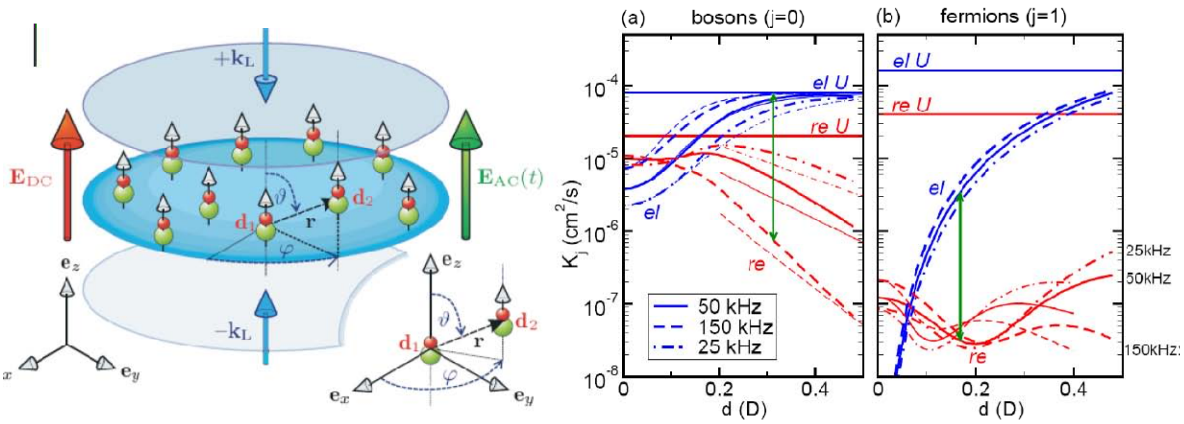

Figure 1. Left: System setup (from [Mic07]): Polar molecules are trapped in the *(x-y)*-plane by an optical lattice made of two counter-propagating laser beams with wave-vectors **KL** (blue arrows). This setup has been now realized experimentally at JILA [deM11]. **Right:** (a) Quasi-2D elastic (el) and reaction (re) rate constants *Kj* for identical KRb (a) bosons and (b) fermions at a collisions energy of E= 240 nK, for three different trap frequencies (25, 50 and 150 kHz) of the confining optical lattice potential, as in the configuration in the left, and the JILA experiment. Vertical arrows indicate a ratio of the elastic to reactive rates of 100, signaling the possibility of performing evaporative cooling of the molecules in these systems (from [Mic10]).

3.1. Universal Rates for Reactive Ultracold Polar Molecules in Reduced Dimensions

In recent work [Mic07,Buc07], we have shown that dipole-dipole interactions between polar molecules can be manipulated using external electric DC, which, possibly in combination with optical trapping to 1D or 2D, allow for the engineering of effective interaction potentials of any given shape between the molecules. This enables a direct *control over elastic and inelastic collisions, and thus over chemical reactions* [Ni10,deM11], in a strongly interacting molecular gas. The conceptually simplest scheme is shown in Fig.1 (left), and corresponds to a system of cold molecules polarized by a DC electric field under conditions of strong transverse confinement, as provided by a deep optical lattice along the *z*-direction [Buc07]. This configuration realizes effective, *purely repulsive,* interactions in 2D with reduced collisional losses, paving the way to the realization of strongly correlated many-body phases in these systems. In recent work [Mic10], we have shown that this setup will strongly reduce molecular reaction rates (that is, collisional losses) in a gas of KRb molecules. Figure (1a) and (b) (right) presents our exact numerical results (continuous lines) for the elastic (el) and reactive (re) rates for bosonic (a) and fermionic (b) molecules, respectively (from [Mic10]). These results show that by increasing the dipole moment *d,* the ratio between elastic and inelastic rates increases significantly for both kinds of molecules, signaling an increased stability of the system. These theoretical studies have been utilized by the JILA group as a guideline for their ground-breaking experimental realization of a 2D quantum gas of fermionic dipoles with reduced collisional losses [deM11].

In addition, in [Mic10] we have shown that the trapping configuration of Fig.1 should also render feasible *evaporative cooling in a gas of KRb molecules*, possibly allowing for the first time reaching quantum degeneracy in cold molecular gases. The arrows in Fig.1 (a) and (b) show parameter regimes where evaporative cooling may be achieved. The JILA group is currently seeking experimental realization of this scheme.

4. New phases of quantum matter with polar molecules

The polar molecule control techniques described in the previous section are expected to lead to the realization of novel many-body phases in the strongly interacting regime with negligible losses, and to applications in the context of quantum simulations. In this funding period, we have worked towards the prediction and understanding of these novel quantum phases that will be studied with the JILA KRb experimental setup or with other promising dipolar systems.

4.1. Quantum phases of cold polar molecules in 2D optical lattices

The addition of an in-plane 2D optical lattice to the configuration of Fig.1 (left) will lead to the realization of strongly correlated phases for arbitrarily weak dipoles. This can be simply understood by considering that in the limit of low molecular densities the main effect of the lattice is to increase the effective mass of the particles, that is, to suppress the kinetic (hopping) energy of the particles. This has the consequence of favoring strong correlations in the system, analogous to cold atomic systems [Jak98].

In [Cap10] we have shown that this 2D lattice configuration will lead to the microscopic realization of Hubbard-like models with long-range interactions, which hold the promise to realize quantum

phases of fundamental theoretical and experimental interest. A key example is the realization of a so-called *supersolid phase*, where solid order coexists with a finite superfluidity in the system. Since its first proposal in the context of the groundstate of Helium, supersolidity has been at the center of interest in condensed matter physics. However, despite a very robust effort both in theory and experiments, its very own existence in physical systems is still a subject of intense debate [Bal10]. In our work in support of the UMd MURI proposal we have studied quantitatively under what conditions interesting quantum phases such as the supersolid phase can appear in a cold ensemble of polar molecules. Using *exact,* state-of-the-art, numerical quantum Monte-Carlo techniques, in [Cap10] we have studied the quantum phases of hard-core bosonic polar molecules on a two-dimensional square lattice interacting via repulsive dipole-dipole interactions. For finite tunneling, we have established the existence of extended regions of parameters where the ground state is a supersolid, obtained by doping the solids either with particles or vacancies (see the phase diagram in Fig.2). In addition, in the limit of small tunneling, we have found clear evidence for a devil's staircase, where Mott solids appear at rational fillings of the lattice. This is an interesting situation which only occurs due to the long-range nature of interactions. In all cases, we have discussed the effects of finite temperature and finite-size confining potentials as relevant to experiments.

This work has already had a strong impact in the community, receiving more than 35 citations in the last year, since its publication (source: http://adsabs.harvard.edu/).

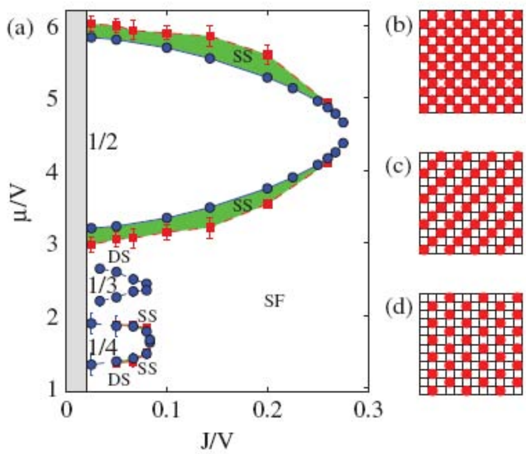

Figure 1: (a) Phase diagram of bosonic polar molecules trapped confined to a 2D optical lattice potential, at temperature T=0. Lobes: Mott solids (densities indicated); SS: supersolid phase; SF: superfluid phase. DS: Devil's staircase. (b-d) Sketches of the ground-state configurations for the Mott solids with filling 1/2, 1/3, and 1/4 respectively. [From Cap10].

4.2. One-Dimensional quantum liquids with power-law interactions: the Luttinger staircase

In [Dal10] we have studied one-dimensional fermionic and bosonic gases with repulsive power-law interactions $1/|x|^B$, with B>1, in the framework of Tomonaga-Luttinger liquid (TLL) theory. These one-dimensional gases will be realized in configurations very similar to the experimental one at JILA [deM11]. In [Dal10] we obtain an accurate analytical expression linking the TLL parameter to the microscopic Hamiltonian, for arbitrary B and strength of the interactions. In the presence of a small periodic potential, power-law interactions make the TLL unstable towards the formation of a cascade of lattice solids with fractional filling, a "Luttinger staircase." Several of these quantum phases and phase transitions are realized with ground state polar molecules and weakly bound magnetic Feshbach molecules.

4.3. Bilayer superfluidity of fermionic polar molecules: Many-Body effects

In [Bar10], we have studied the BCS superfluid transition in a single-component fermionic gas of dipolar particles loaded in a tight bilayer trap, with the electric dipole moments polarized perpendicular to the layers. This corresponds to the configuration of Fig.1 (left), when more than one layer of the optical lattice is populated, as it happens in experiments [deM11]. Based on the detailed analysis of the interlayer scattering, we have calculated the critical temperature of the interlayer superfluid pairing transition when the layer separation is both smaller (dilute regime) and on the order or larger (dense regime) than the mean interparticle separation in each layer. Our calculations go beyond the standard BCS approach and include the many-body contributions resulting in the mass renormalization, as well as additional contributions to the pairing interaction. We find that the many-body effects have a pronounced effect on the critical temperature and can either decrease (in the very dilute limit) or increase (in the dense and moderately dilute limits) the transition temperature as compared to the BCS approach. These results have direct relevance to the experiments at JILA.

5. Novel dipolar systems for quantum simulations: Rydberg-dressed atoms

In recent work [Pup10,Cin10], we have opened a new line of research in the study of dipolar gases by proposing ways to obtain long-range interactions in quantum gases of ground-state atoms [Pup10, Sch10], by weakly-dressing the atoms with excited electronic Rydberg states with a large dipole moment, using laser light. Since in this situation spontaneous emission from the excited atomic states can be largely suppressed, these techniques open the way to engineering long-range interactions and strongly correlated phases in atomic gases, with reduced decoherence from spontaneous emission and inelastic collisions.

5.1 Strongly correlated gases of Rydberg-dressed atoms: quantum and classical dynamics

In [Pup10] we discuss techniques to generate long-range interactions in a gas of ground state alkali atoms, by weakly admixing excited Rydberg states with laser light. This provides a tool to engineer strongly correlated phases with reduced decoherence from inelastic collisions and spontaneous emission. As an illustration, we discuss the quantum phases of dressed atoms with dipole-dipole interactions confined in a harmonic potential, as relevant to experiments. We show that residual spontaneous emission from the Rydberg state acts as a heating mechanism, leading to a quantum-classical crossover.

5.2 Supersolid Droplet Crystal in a Dipole-Blockaded Gas

In [Cin10] we utilize the techniques developed in [Pup10] for generating long-range interactions in cold gases of alkali atoms in order to study new exotic quantum phases. In particular, in [Cin10] a novel supersolid phase is predicted for an ensemble of Rydberg atoms in the dipole-blockade regime, interacting via a repulsive dipolar potential softened at short distances. A supersolid phase is the coexistence of superfluid and solid behaviour, a “Holy-Grail” of condensed-matter physics, long-

sought-for in the context of the groundstate of He4 [Bal10] and never observed before. Different from, e.g. [Cap10], here the supersolid is *self-assembled,* meaning that it appears in *free-space,* even in the absence of an underlying optical lattice potential.

Using exact numerical techniques, in [Cin10] we have studied the low-temperature phase diagram of this system, and have observed an intriguing phase consisting of a crystal of mesoscopic superfluid droplets. At low temperature, phase coherence throughout the whole system, and the ensuing bulk superfluidity, are established through tunnelling of identical particles between neighboring droplets, giving rise to a supersolid in free-space.

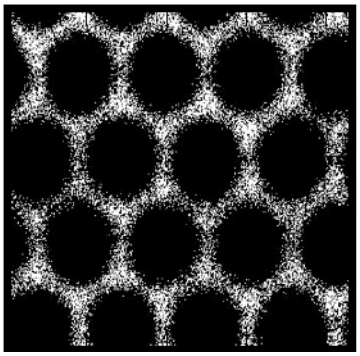

Figure 3: Quantum Monte-Carlo snapshot of a 2D supersolid in free space, made of 200 Rydberg-dressed atoms [Cin10]. Darker regions reflect larger densities.

5.3 Dynamical crystal creation with polar molecules of Rydberg atoms in optical lattices

In [Sch10] we investigate the dynamical formation of crystalline states with systems of polar molecules or Rydberg atoms loaded into a deep optical lattice. External fields in these systems can be used to couple the atoms or molecules between two internal states: one that is weakly interacting and one that exhibits a strong dipole-dipole interaction. By appropriate time variation of the external fields, we show that it is possible to produce crystalline states of the strongly interacting states with high filling fractions chosen via the parameters of the coupling. We study the coherent dynamics of this process in one dimension (1D) using a modified form of the time-evolving block decimation (TEBD) algorithm, and obtain crystalline states for system sizes and parameters corresponding to realistic experimental configurations. For polar molecules these crystalline states will be long-lived, assisting in a characterization of the state via the measurement of correlation functions. We also show that as the coupling strength increases in the model, the crystalline order is broken. This is characterized in 1D by a change in density-density correlation functions, which decay to a constant in the crystalline regime, but show different regions of exponential and algebraic decay for larger coupling strengths.

6. Conclusions

In this first funding period we have obtained several key results towards the control of cold collisions between molecules, and the prediction and understanding of exotic many-body phases of ultracold molecules trapped in optical lattices. Highlights are: (a) The computation of the elastic and reactive rates of quasi-two-dimensional and quasi one-dimensional collisions of highly reactive ultracold molecules (in collaboration with the UMd MURI PI Paul Julienne). Some of these predictions have been now verified experimentally by the JILA group [deM11]. (b) The determination of the phase diagram of ultracold polar molecules trapped in optical lattices in two [Cap10] and one [Dal10] dimensions, showing that the groundstate of the system can support exotic phases such as supersolid phases and Devil-like's staircases of lattice solids [Cap10, Dal10]. (c) The study of the physics of dipolar gases as can be realized in novel setups, such as quantum gases of laser-dressed

atoms [Pup10, Sch10]. These works have already resulted in several high-profile publications in this first funding period. Future work will focus on the analysis of the quantum phases of bosonic and fermionic polar molecules in multi-layer structured, as are being realized in experiments with molecules trapped in several layers of an optical lattice. We expect that several interesting pairing mechanisms will arise in these systems, possibly leading to many-body phases of interest. In addition, we will work towards the realization of new architectures for quantum computing with polar molecules which make specific use of long-range interactions in these systems.

7. Published papers within EOARD (first funding period)

1) F. Cinti, P. Jain, M. Boninsegni, A. Micheli, P. Zoller, and G. Pupillo, *Supersolid Droplet Crystal in a Dipole-Blockaded Gas,* Phys. Rev. Lett. **105**, 135301 (2010).

2) B. Capogrosso-Sansone, C. Trefzger, M. Lewenstein, P. Zoller, and G. Pupillo, *Quantum Phases of Cold Polar Molecules in 2D Optical Lattices*, Phys. Rev. Lett. **104**, 125301 (2010).

3) M. Dalmonte, G. Pupillo, and P. Zoller, *One-Dimensional Quantum Liquids with Power-Law Interactions: The Luttinger Staircase,* Phys. Rev. Lett. **105**, 140401 (2010).

4) A. Micheli, Z. Idziaszek, G. Pupillo, M. A. Baranov, P. Zoller, and P. S. Julienne, *Universal Rates for Reactive Ultracold Polar Molecules in Reduced Dimensions*, Phys. Rev. Lett. **105**, 073202 (2010).

5) G. Pupillo, A. Micheli, M. Boninsegni, I. Lesanovsky, and P. Zoller, *Strongly correlated gases of Ryydberg-dressed atoms: quantum and classical dynamics,* Phys. Rev. Lett. **104**, 223002 (2010).

6) J. Schachenmayer, I. Lesanovsky, A. Micheli, and A.J. Daley, *Dynamical crystal creation with polar molecules of Rydberg atoms in optical lattices,* New J. Phys. **12**, 103044 (2010).

7) M. Baranov, A. Micheli, S. Ronen, and P. Zoller, *Bilayer superfluidity of fermionic polar molecules: Many-Body effects,* Phys. Rev. A **83**, 043602 (2011).

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[Dal10] M. Dalmonte, G. Pupillo, and P. Zoller, *One-Dimensional Quantum Liquids with Power-Law Interactions: The Luttinger Staircase,* Phys. Rev. Lett. **105**, 140401 (2010).

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[Sch10] J. Schachenmayer, I. Lesanovsky, A. Micheli, and A.J. Daley, *Dynamical crystal creation with polar molecules of Rydberg atoms in optical lattices,* New J. Phys. **12**, 103044 (2010).

9. List of Symbols, Abbreviations, and Acronyms

JILA group: Debbie Jin's and Jun Ye's experimental group at http://jila.colorado.edu/